Atomic Engly Act 195

C66-2146



WANL-TNR-167

July, 1964

NASA CR 70 7 4 2

TO UNCLASSIFIED TO UNCLASSIFIED TO UNCLASSIFIED TO THE STREET TO THE STR

PREPARED BY:

L. Cadoff,

L. Fleischer, W. Brizes Automana Declassification

AUTHORIZED CLASSIFIER

INFORMATION CATEGORY

Confidential-Restricted Data

STATES TO STATE ESPIONAGE LAWS TRIES

APPROVED BY:

J. Martin Tobin, Supr.
Basic Fuel Studies
Materials Department

a. Boltay

A. Boltax, Mgr.
Fuel Development
Materials Department

ANALYSIS OF ALPHA
PARTICLE RELEASE FROM
PARTICULATE FUEL

DATE

(Title Unclassified)





# Table of Contents

			Page	
Abstract				
1.	Introduction			
П.	Analytical Model for Alpha Particle Release			
ш.	Disc	Discussion of Analytical Results		
IV.	Experimental Results and Discussion			
٧.	Con	Conclusions		
VI.	Ack	nowledgment	edgment 10	
VII.	Appendix			
	A.	Development of a Generalized Alpha Escape Equation	. 11	
	В.	Derivation of Alpha Equation Assuming Variable Range	16	
	c.	Derivation of Alpha Equation Assuming a Constant Range	22	



stronuclear
WANL-TNR-167
July, 1964

#### **Abstract**

50472

Mathematical expressions are presented for relating experimental alpha count data with the extent of migration in pyrocarbon coated UC<sub>2</sub> particles. The expressions are based on a simplified model for migration in which the uranium concentration profile in the migration zone is approximated by a step function. Experimental data are shown to agree satisfactorily with the derived equations.

It is concluded that alpha counting techniques are primarily applicable, in conjunction with microradiography, to determine the uranium concentration in the migration zone and the total weight of migrated uranium.







#### Introduction

Alpha count techniques have been employed by WANL and other laboratories to study uranium migration in pyrocarbon coated,  $UC_2$  fuel particles. The experimental alpha count data, however, has in general been difficult to interpret, showing only limited agreement with results obtained independently, by methods such as microradiography. This apparent inconsistency has raised doubts as to the validity of the alpha count technique. The purpose of the present study was to investigate the use of alpha counting with respect to the study of migration phenomena.

### II. Analytical Model for Alpha Particle Release

Figure 1 shows a typical pyrocarbon coated UC<sub>2</sub> spherule in which uranium has migrated into the pyrocarbon coat. That annular region of the pyrocarbon coating into which the uranium has penetrated is designated the migration zone. The alpha particles are generated in this zone by the decay of the uranium radioisotopes, the rate of release being dependent on their decay rate (activity) and concentration. A given uranium nucleus has an equal probability of emitting an alpha in any direction. The range of the emitted alpha particle increases with increasing energy and decreases with increasing electron density of the medium through which it passes. Only if the alpha is not deenergized and halted by the intervening material will it escape from the fuel particle and be detectable.

The number of detectable alpha particles per unit time, originating at a point in the migration zone can be expressed as the product of the rate of alpha decay and the probability of an alpha generated at that point escaping from the fuel particle. It is clear, therefore, that the total alpha escape rate per fuel particle will be the integral of this product over all points in the migration zone.

Employing the above principle, two analytical expressions have been derived (see Appendix) to correlate alpha count data to the extent of fuel migration in pyrocarbon coated fuel particles. The basic assumptions in the analysis are:







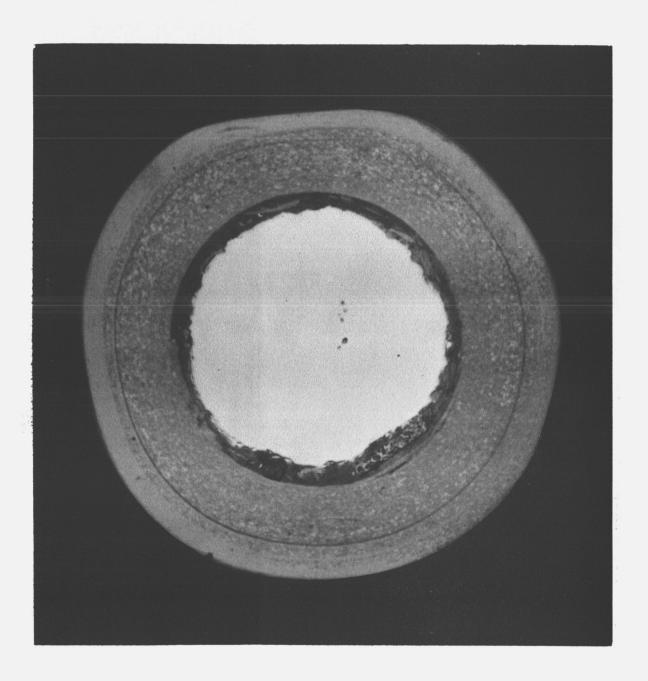


Figure 1 - Photomicrograph of Coated Fuel Particle with Migration Zone







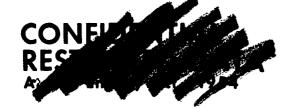
- 1. Spherical core and particle geometry.
- 2. A spherical migration zone moving out symmetrically from the core into the pyrocarbon.
- 3. A uniform uranium concentration, homogeneously distributed in the migration zone.
- 4. A discontinuous drop in the uranium concentration to zero at the leading edge of the migration front "T". This approximation is an attempt to describe the metallographic and radiographic observations of a sharp demarcation between the migration zone and the uneffected PyC coating.
- 5. Only alpha particles generated in the migration zone can escape from the fuel particle. This approximation requires that the coating thickness must be equal to or greater than the range of the alpha in the pyrocarbon.
- 6. The alpha energy can be approximated by the weighted mean energy from all emitting radioisotopes. (For enriched uranium, the average alpha energy is 4.75 MeV.)

# III. Discussion of Analytical Results

Equations 20 and 21 which appear in the Appendix, relate experimental alpha counting rate data to the radial extent of migration, "T", for a single fuel particle. Equation 20 takes into account the variation of alpha particle range with uranium concentration through which the alpha passes on its escape path. This expression is quite cumbersome and has been programmed for the IBM-7094 computer. Equation 21 was derived by making the simplifying assumption that for dilute uranium concentrations in the migration zone, the range can be considered to be constant and equivalent to the alpha range in pyrocarbon.

Figure 2 shows the relationship of alpha escape rate, per coated fuel particle (Equations 20 and 21) as a function of the radial extent of migration, "T". The values shown were calculated using nominal fuel particle parameters.\* It can be seen that

<sup>\*</sup> e.g., R = 75 $\mu$ , r = 12.55 mg/cm<sup>2</sup>, r = 3.85 mg/cm<sup>2</sup> (alpha energy = 4.75 MeV for enriched uranium),  $\rho_c = 1.6$  g/cm<sup>3</sup>, r' = 24.1 $\mu$ ,  $\lambda = 133$  dpm/ $\mu$  gm, W = 0.20,  $S^d = 1$ . The symbols used are defined under Equations 20 and 21 in the appendix.



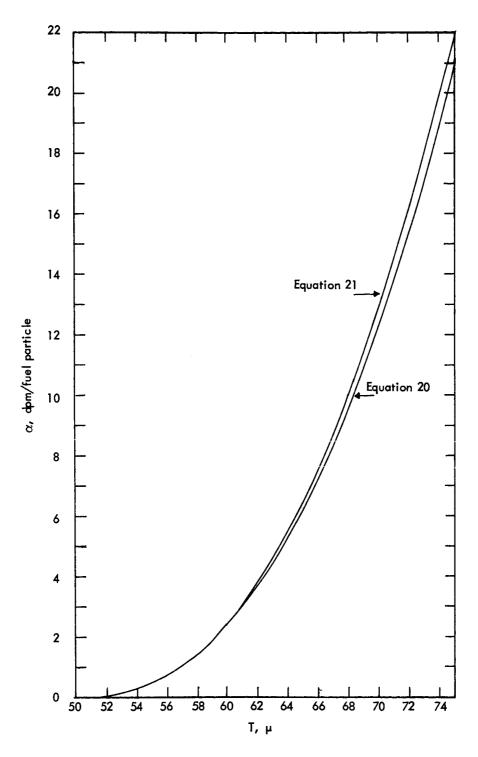


Figure 2 - Alpha Counts Emitted vs. Radial Extent of Migration (UC $_2$  core radius = 50 $\mu$ , pyrocarbon coating thickness = 25 $\mu$ )







good correlation is obtained between Equations 20 and 21. The two expressions are nearly identical at low values of "T", the radial extent of migration. The alpha particle encounters only a small region of uranium bearing pyrocarbon (migration zone) on its escape path and the alpha range is, therefore, nearly equivalent to its range in pyrocarbon. However, as the value of "T" approaches "R" (the radius of the fuel particle), the curve describing Equation 21 tends to diverge in a positive direction from Equation 20. This is because Equation 21 assumes that the alpha escape range is equal to the range in pyrocarbon, while Equation 20 accounts for the small reduction in range of the alpha particle as it passes through the migration zone. Thus, it is expected that Equation 21 will slightly overestimate the rate of alpha release for particles which exhibit extensive amounts of migration.

Ideally, if one knows the concentration of uranium, "W", in the migration zone the radial extent of migration, "T", can be obtained for a single fuel particle from the measured rate of alpha counts. Conversely, if the extent of migration and the rate of alpha release is measured, the value of  $W_U$  can be determined. Equation 21 permits explicit determination of  $W_U$ , whereas if equation 20 is used, the weight fraction of uranium in the migration zone must be determined by methods of approximation.

The problem of correlating the extent of migration with alpha count data becomes more complex when we deal with a large number of coated particles. It is known that heat-treating a sample batch of fuel particles results in a distribution of the extent of migration among the particles. As can be seen from Figure 2, alpha particles emitted from a few completely migrated fuel particles can heavily outweigh those emitted from many lesser migrated fuel particles. In addition, the number of alpha particles emitted is not a linear function of extent of migration. It can, therefore, be concluded that, in general, no correlation should be expected between the measured number of alpha particles and the extent of migration that has been averaged over all fuel particles.







To demonstrate the validity of Equations 20 and 21, it is necessary to determine the distribution of migration in a sample of heat-treated fuel particles. The total number of alpha particles emitted by this sample per minute would then be the weighted sum of the product of the distribution function and Equations 20 or 21.

#### IV. Experimental Results and Discussion

Samples of NUMEC and 3M enriched fuel particles (nominal radius =  $75\mu$ ) were heat treated for various temperatures and times as indicated in Table 1. Alpha count measurements were made on 0.05 gm samples using a Nuclear Measurements Corporation flow counter with P-10 gas. Estimated counting efficiency is 0.30. Measurements of extent of migration were originally made from microradiographs of 100 particles from each sample. It was later found that 50 particles were sufficient to give good statistics for this analysis.

Using the measured migration distribution and the experimental rate of alpha release obtained for both NUMEC and 3M particles at  $2300^{\circ}$ C, values of W<sub>U</sub>, the weight fraction of uranium in the migration zones were determined from Equations 20 and 21. Nearly equivalent values of W<sub>U</sub> were obtained using either equation and the mean values are presented in Table 1.

The calculated values of  $W_U$ , obtained on  $2300^{\circ}C$  and the measured distribution functions determined for all other temperatures were then applied to Equation 20, and the rate of detectable alpha release computed. The results are given in Table 1. For purposes of comparison, the calculated rate of alpha release based on the previously determined values of  $W_U$  and the extent of migration averaged over all particles are also shown.

Table 2 indicates that, in general, at temperatures below  $2400^{\circ}$ C good correlation is achieved between experimental alpha values and those calculated on the basis of the distribution function. This agreement was obtained by applying the calculated values of the uranium concentration in the migration zone (W = 0.21 and 0.11), determined at  $2300^{\circ}$ C for NUMEC and 3M particles, respectively, to all other temperatures. It appears, therefore, that the uranium concentration in the migration zone, for







Table 1 - Comparison of Experimental and Calculated Values of Alpha Counts on PyC Coated Fuel Particles

Supplier	Heat-Treatment Schedule	Wt. Fraction*	Experimental αcounts/min-gm	Calculated <sup>(1)</sup> <sup>α</sup> counts∕min–gm	Calculated <sup>(2)</sup> <sup> α</sup> counts√min–gm
NUMEC	As received		3,000		
	2000°C, 1 hr.		36,800	21,800	8, 200
	2100°C, 1 hr.		34,500	27, 200	8, 200
	2200°C, 1 hr.		48,300	56,700	19,500
	2300°C, 1 hr.	0.21	57,200	57, 200	19,500
	2400°C, 1 hr.		1,200,000	286, 000	581,000
3M	As received		3,600		
	2100°C, 6 hr.		84,250	98,000	80, 500
	2200°C, 6 hr.		149,400	148,000	118,000
	2300°C, 6 hr.	0.11	153,000	153,000	118,000

<sup>(1)</sup> Based on migration distribution function and Equation 20.

<sup>(2)</sup> Based on averaged extent of migration

<sup>\*</sup> Weight fraction of uranium in migration zone.





any given sample of material is reasonably constant over a wide range of temperatures. Electron beam microprobe data (see Table 2) obtained for the same lot of NUMEC material used in this study, confirms that W<sub>U</sub> is approximately 0.21 and that it is relatively independent of temperature of heat treatment. Microprobe data is not yet available on the uranium concentration in the 3M particles.

The computed rate of alpha release determined from the values of the extent of migration averaged over all particles do not, in general, adequately agree with the experimental data. The poor correlation with respect to the NUMEC particles can be traced to the non-linear relationship which exists between alpha release rate and the extent of migration (Figure 2), and to the wide distribution of migration exhibited by these particles. The migration distribution functions were considerably narrower at all temperatures for the 3M particles, resulting in a somewhat better agreement with experimental alpha release values.

At 2400°C, the migration behavior in many particles is inhomogeneous. The migration zone is comprised of large discrete granules of UC<sub>2</sub> embedded in the PyC coat. It is apparent that Equations 20 and 21, which assume a uniform uranium concentration in the migration zone are not applicable for this type of behavior.

### V. Conclusions

Two equations are presented to describe the relationship between alpha particle release from a single coated fuel particle and the radial extent of fuel migration. These equations can be applied to a large sample of fuel particles to determine the total rate of alpha release if the extent of migration in all particles (migration distribution) is known. The agreement obtained between experimental and calculated alpha count values indicates that the assumptions made in this analysis are reasonable.

It is considered that alpha count measurements on a large sample of fuel particles will not provide an effective measure of the average extent of migration. This is because the alpha values obtained are strongly dependent on the migration distribution in the sample. Experimental results obtained on fuel particles from differing sample lots or







Table 2 - Summary of Uranium Concentrations Found in Migration Zones of Loose Enriched NUMEC Coated Particles Obtained from Electron Beam Microprobe Examination

Time and Temperature Heat Treatment	Weight Fraction Uranium in Migration Zone		
1900°C - 9 hours	0.16		
1900°C - 9 hours	0.21		
2000°C - 21 hours	0.19		
2100°C - 9 hours	0.22		
2200°C - 9 hours	0.20		
2300°C - 0.5 hours	0.22		
2400°C - 0.5 hours	0.22		
2400°C - 0.5 hours	0.10 to 0.83*		

<sup>\*</sup> Completely migrated particle exhibiting inhomogeneous migration.





suppliers can also lead to misleading conclusions as to the extent of migration because of possible differences in the uranium concentrations in the migration zone.

Alpha count measurements, combined with the experimentally determined migration distribution function, may permit determination of the concentration of uranium in the migration zone of coated fuel particles. Obviously, the total weight of migrated uranium can be calculated from this concentration and the assumed geometry. This is perhaps the most promising feature of the alpha count technique. Together with parametric studies of pyrocarbon coating density, structure and orientation, the uranium concentration values obtained by the alpha count method may provide basic insight into the fuel migration phenomenon.

#### VI. Acknowledgment

The authors would like to thank J. M. Tobin and D. C. Jacobs for their helpful comments and discussions during the course of this study.



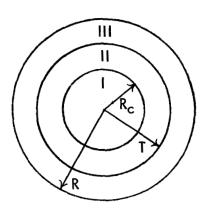




# VII. Appendix

# A. Development of a Generalized Alpha Escape Equation

Figure 3 illustrates the assumptions made in establishing a model to correlate alpha count data to the extent of fuel migration.



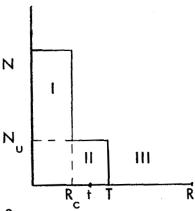


Figure 3

R = radius of fuel particle, microns;  $R_c$  = radius of fuel core, microns; t = radial position variable; T = radial extent of migration zone, microns; N = uranium concentration,  $\mu g/\mu^3$ ;  $N_u$  = uranium concentration in migration zone,  $\mu g/\mu^3$ , where  $N_u$  can be approximated as:

$$N_{U} = \frac{W_{U} p_{U} p_{c}}{W_{U} (p_{c} - p_{U}) + p_{U}} \times 10^{-6}$$

 $10^{-6}$  = factor to give N<sub>U</sub> in units of  $\mu g/\mu^3$ ; p<sub>U</sub> = density of uranium,  $g/cm^3$ ; p<sub>C</sub> = density of pyrocarbon coat,  $g/cm^3$ ; W<sub>U</sub> = weight fraction of uranium in migration zone.

The assumptions for this model are:

- 1) A spherical core and particle geometry (Regions I and III)
- 2) A spherically symmetrical migration zone (Region II)







- 3) A uniform uranium concentration, homogeneously distributed in the migration zone (Region II).
- 4) A discontinuous change in uranium concentration from  $N_{_{\mbox{U}}}$  to zero at point "T", the radial extent of migration.
  - 5) All escaping alpha's are born in the migration zone (Region II).
  - 6) The emitted alpha particles are monoenergetic.

Referring to Figure 3, the rate of escaping alpha's, originating at a point "t" in the migration zone, is the product of the rate of alpha emission at that point and the geometrical probability of escape from that point. Analytically,

$$d\emptyset(t) = P(t) \lambda dB(t)$$
 (1)

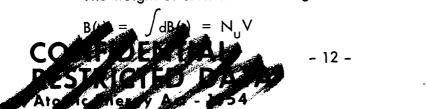
when  $d\emptyset(t)$  = rate of alpha escape from any volume element centered about radial position, "t"; P(t) = geometric probability of escape from any radial point, "t";  $\lambda$  = average specific alpha activity of uranium; dB(t) = weight of uranium in a volume element centered about point "t". Integrating Equation 1 over the migration zone yields, " $\alpha$ ", the total rate of alpha escape per particle.

$$\alpha = \lambda \int_{t'}^{T} P(t) dB(t) = \lambda \int_{t'}^{T} P(t) \partial B/\partial t \quad dt \text{ (since B = f(t) only)}$$
 (2)

The upper limit of integration, "T", is fixed by the radial extent of migration, -- beyond this point dB(t) = 0. The lower limit, "t' " refers to some point in the migration zone where the alpha range is just sufficient to reach the surface of the fuel particle.
"t' " will be evaluated later in this report.

The individual terms of Equation 2, (P(t)) and B(t) are derived in the following sections.

Weight of Uranium in Migration Zone, B(t)
 The weight of uranium in the migration zone of a fuel particle is:







where V = volume of material in migration zone, bounded by point "t"

$$V = 4/3\pi (t^3 - R_c^3)$$
 (4)

Therefore,

$$B(t) = N_{U} (4/3 \pi) (t^{3} - R_{c}^{3})$$
 (5)

and

$$\partial B/\partial t = N_{U} (4\pi t^{2}) \tag{6}$$

### 2) Geometric Probability of Alpha Escape, P(t)

An alpha particle, generated at point "t" in the migration zone will escape from the fuel particle only if its range in the intervening material is equal to or greater than the distance from its point of origin to the surface of the fuel particle. Since the emitted alpha has an equal probability of traveling in any direction, one can represent the alpha emittance by a sphere of radius,  $r_{\alpha}$  where  $r_{\alpha}$  is the range of an escaping alpha particle. The solid angle of emittance, therefore  $r_{\alpha} = 4\pi$ . See Figure 4.

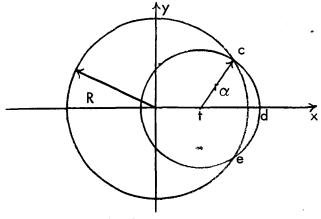


Figure 4

The alpha escape volume is defined by the solid angle " $\Omega$ ", subtended by the intersection of the emittance sphere ( $r_{\alpha}$ ) with the fuel particle sphere (R). It, thus, follows that the probability of alpha escape, P (t) is





The solid angle of escape, "  $\Omega$ ", can be obtained from the surface area of the emittance sphere, S which lies outside the fuel particle.

The surface area  $S_{ce}$  is determined by rotation of segment cd about the x axis by the following standard equation:

$$S_{ce} = 2\pi \int_{C}^{d} y \left[ 1 + (\lambda y/\lambda x)^{2} \right]^{1/2} dx$$
 (8)

where  $y = \left[ r_{\alpha}^2 - (x-t)^2 \right]^{1/2}$ 

$$c = \frac{R^2 - r_{\alpha}^2 + t^2}{2t}$$

$$d = t + r_{\alpha}$$

Substitution and integration of Equation 8 yields

$$S_{ce} = \pi r_{\alpha} \left[ \frac{\left(r_{\alpha} + t\right)^{2} - R^{2}}{t} \right]$$
 (9)

For a spherical system

$$\Omega = \frac{S_{ce}}{r_{co}^2} = \frac{\pi \left[ (r_{co} + t)^2 - R^2 \right]}{r_{co} t}$$
 (10)

Substituting into Equation 7 gives the geometric probability of escape;

$$P(t) = \Omega/4 \pi = \frac{(r_{\alpha} + t)^2 - R^2}{4 r_{\alpha} t}$$
 (11)

3) Final Form of General Alpha Equation

Substituting Equation 6 into Equation 2 gives
$$\alpha = 4 \pi N_{U} \lambda \int_{t}^{T} P(t) t^{2} dt$$
(12)







To account for experimental counting efficiency, a factor "  $\epsilon$  " is inserted into Equation 12.

$$\alpha_{\alpha} = 4 \pi N_{u} \lambda \in \int_{t^{1}}^{T} P(t) t^{2} dt$$
 (13)

where  $\alpha_{a}$  = experimentally determined rate of alpha escape per fuel particle.

Finally, by substituting Equation 11 into Equation 13 we obtain the generalized equation to relate alpha count data with extent of migration:

$$\alpha_{\alpha} = 4 \pi N_{U} \lambda \in \int_{t'}^{T} \left[ \frac{(r_{\alpha} + t)^{2} - R^{2}}{4 r_{\alpha} t} \right] t^{2} dt$$
 (14)





## B. Derivation of Alpha Equation Assuming Variable Range

In this section, Equation 14 will be solved by taking into account, the variation of alpha range "r", with position "t" in the migration zone.

In a homogeneous two component system, (e.g., U and C) "r", the range of an alpha particle of given energy depends on the relative amounts of the components in the mixture and the ranges in the individual components. Bragg has developed the following formula to approximate the range, "r", of an alpha particle in a two component system:

$$r = \left(\frac{r_{u} r_{c}}{W_{u} (r_{c} - r_{u}) + r_{u}}\right) \left(\frac{10}{p_{mix}}\right)$$
 (15)

where  $r_U$  = thickness density for an alpha in U, mg/cm<sup>2</sup>;  $r_C$  = thickness density for an alpha in C;  $W_U$  = weight fraction of uranium in mixture;  $p_{mix}$  = density of the homogeneous mixture of U and C through which the alpha passes, g/cm<sup>3</sup>; and

$$p_{mix} = \frac{p_U p_C}{W_U (p_C - p_U) + p_U}$$

 $P_{U}, P_{C} = \text{density of U and C respectively, g/cm}^{3}$ . The factor 10 converts r to units of microns.

In the model proposed in this analysis, a detectable alpha particle must pass through two discrete regions of different composition (see Figure 5). The alpha originates in the migration zone (Region II), a dilute, uniform dispersion of uranium in carbon (weight fraction, W<sub>U</sub>). Since the size of the dispersed phase is small with respect to "r", Region II is considered to be homogeneous. The alpha particle passes from the migration zone into the uneffected pyrocarbon coating, Region III.

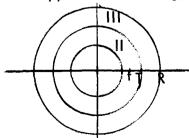


Figure 5





The Bragg relation, Equation 15, is not strictly applicable to this heterogeneous system because the  $\alpha$  energy is attenuated at different rates in each region. However, if the atomic concentration of U in the migration zone is low (e.g.,  $W_U=0.2$  or a/o U=1.26), the heterogeneous system can be approximated by a homogeneous system containing the same amount of uranium. That is, the uranium in the migration zone is taken, for purposes of this approximation, to be spread uniformly through both the migration zone and the uneffected coating. Thus, the uranium concentration becomes the mean concentration from the alpha particle's point of origin to the surface of the fuel particle. This mean concentration is defined as:

$$\frac{1}{W_{U}} = \frac{\int_{t}^{T} \frac{W}{V} \frac{dt}{dt}}{\int_{t}^{R} \frac{R}{dt} \frac{dt}{dt}} = \frac{W}{R} \frac{(T-t)}{-t}$$
(16)

Therefore, the approximate alpha range is given by:

$$r_{\alpha} = \frac{r_{u} r_{c}}{\overline{W_{u}} (r_{c} - r_{u}) + r_{u}} \times \frac{10}{\overline{P_{mix}}}$$

$$(17)$$

where

$$\frac{P_{\text{mix}}}{P_{\text{mix}}} = \frac{P_{\text{u}} P_{\text{c}}}{W_{\text{u}} (P_{\text{c}} - P_{\text{u}}) + P_{\text{u}}}$$

Implicit in Equation 16 is that the composition along the limiting escape path (the path which defines solid angle " $\Omega$ ") can be approximated by the composition along the radial escape path,  $\frac{1}{W_U}$ . This approximation has been tested by numerical methods and very satisfactorily represents the actual case.

Substitution of Equation 16 into Equation 17 yields,

$$r_{\alpha} = \frac{10 r_{u} r_{c}}{p_{u} p_{c}} \left( \frac{L - St}{M - Qt} \right)$$
 (18)







where

$$L = W_U (p_C - p_U)T + p_U R$$

$$S = W_U (p_C - p_U) + p_U$$

$$Q = W_{U} (r_{c} - r_{U}) + r_{U}$$

$$M = W_{U} (r_{C} - r_{U})T + r_{U}R$$

L, S, Q, and M are arbitrary factors to simplify Equation 18.

As previously mentioned, the lower limit of integration, "t' ", of Equation 14 is that position in the migration zone where the radial range will be sufficient to just reach the fuel particle surface. This range is defined as "r " and occurs at position:

$$t = t^1 = R - r_0$$

 $r_{\alpha}$  is evaluated by substituting into Equation 18 the limits that  $r_{\alpha} = r_{\alpha}$  when

$$t = R - r_0$$
. Simplifying,

$$r_{o} = -\left[M - QR - \frac{10 r_{u}r_{c}}{P_{u}P_{c}}S\right] + \left[\left(M - QR - \frac{10 r_{u}r_{c}}{P_{u}P_{c}}S\right)^{2} + 2 Q\right]$$

$$\frac{40 r_{0} r_{c}}{P_{0} P_{c}} Q (L - SR)$$
(19)

The expression for  $r_{\alpha}$  (Equation 18) can now be applied to Equation 14. Integration and substitution of the limit  $t' = R - r_{o}$ , yields  $\alpha_{a}$ , the actual rate of alpha release from a single fuel particle:





$$\alpha_{a} = 4\pi \lambda N_{u} \in \left[ \frac{p_{u} p_{c}}{40 r_{u} r_{c}} \left[ \left[ (L - ST) - (L - S(R - r_{o})) \right] \left[ - \frac{R^{2}M}{S^{2}} + \frac{2LQR^{2}}{S^{3}} + \right] \right] \right]$$

$$\frac{3 L^{2}M}{S^{4}} - \frac{4 L^{3}Q}{S^{5}} \right] + \left[ (L - ST)^{2} - (L - S(R - r_{o}))^{2} \right] \left[ -\frac{3LM}{2S^{4}} - \frac{QR^{2}}{2S^{3}} + \frac{3 L^{2}Q}{S^{5}} \right] +$$

$$\left[ (L - ST)^{3} - (L - S(R - r_{o}))^{3} \right] \left[ \frac{M}{3S^{4}} - \frac{4QL}{3S^{5}} \right] + \left[ (L - ST)^{4} - (L - S(R - r_{o}))^{4} \right]$$

$$\left[\frac{Q}{4 \, S^{5}}\right] + \ln \frac{L - ST}{L - S(R - r_{o})} \left[ + \frac{R^{2} \, M \, L}{S^{2}} - \frac{Q \, R^{2} L^{2}}{S^{3}} - \frac{M \, L^{3}}{S^{4}} + \frac{Q \, L^{4}}{S^{5}} \right] \right] +$$

$$\frac{T^{3}-(R-r_{o})^{3}}{6}+\frac{10\,r_{u}\,r_{c}}{4\,p_{u}\,p_{c}} \qquad \left[\ln \frac{M-Q\,T}{M-Q\,(R-r_{o})}\,\left[\left(-\frac{M\,L}{Q^{2}}+\frac{M^{2}S}{Q^{3}}\right)\right]\right. +$$

$$\left[ (M - QT) - (M - Q(R - r_0)) \right] \left[ \frac{L}{Q^2} - \frac{2MS}{Q^3} \right] +$$

$$\left[ (M - QT)^{2} - (M - Q(R - r_{o}))^{2} \right] \left[ \frac{s}{2Q^{3}} \right]$$
(20)





where:

$$r_{o} = \frac{-\left[M - QR - \frac{10 r_{u} r_{c}}{P_{u} P_{c}} S\right] + \left[\left(M - QR - \frac{10 r_{u} r_{c}}{P_{u} P_{c}} S\right)^{2} + \frac{2 Q}{P_{u} P_{c}}\right]}{2 Q}$$

$$L = W_{U} (p_{c} - p_{U})T + p_{U}R$$

$$S = W_U (p_C - p_U) + p_U$$

$$Q = W_U (r_C - r_U) + r_U$$

$$M = W_{U} (r_{c} - r_{U})T + r_{U}R$$

and:

 $\alpha_{\rm a}$  = Total number of experimental alpha counts, dpm/fuel particle.

 $\lambda$  = Specific alpha activity of uranium, dpm/ $\mu$ g.

 $N_{\perp}$  = Concentration of uranium in migration zone,  $\mu g/\mu^3$ 

$$N_{\rm u} = \frac{W_{\rm u} p_{\rm c} p_{\rm u}}{W_{\rm u} (p_{\rm c} - p_{\rm u}) + p_{\rm u}} \times 10^{-6}$$

W = Weight fraction of uranium in migration zone.

 $P_{..}$  = Density of uranium, g/cm<sup>3</sup>.

 $p_{c}$  = Density of pyrocarbon coat, g/cm<sup>3</sup>.

 $\epsilon$  = Experimental counting efficiency factor.

Thickness density for an alpha particle in uranium, mg/cm<sup>2</sup>.





 $r_c = Thickness density for an alpha particle in carbon, mg/cm<sup>2</sup>.$ 

R = Radius of coated fuel particle,  $\mu$ .

T = Radial extent of migration,  $\mu$ .





# C. Derivation of Alpha Equation Assuming a Constant Range

The functional relationship between the rate of alpha release from a fuel particle and the extent of fuel migration can be greatly simplified if it is assumed that the range of an emitted alpha is constant and independent of position of origin in the migration zone. This assumption is valid for low uranium concentrations in the migration zone. For example, the range of a 4.75 MeV alpha in a homogeneous mixture of uranium and carbon ( $W_U = 0.2$ ) is 22.8 microns. This compares to a range of about 24.1 microns in 1.6 g/cm<sup>3</sup> dense carbon.

In this derivation, we set the alpha escape range equal to  $r'_c$ , the range of an alpha in pyrocarbon. Substituting " $r'_c$ " into Equation 14, and setting  $t' = R - r'_c$ , one obtains after integration, the following equation to relate alpha release to migration.

$$\alpha_{\alpha} = 4 \pi \lambda N_{U} \epsilon \left[ \frac{(r_{c}')^{2} - R^{2}}{8 r_{c}'} (T^{2} - (R - r_{c}')^{2}) + \frac{T^{3} - (R - r_{c}')^{3}}{6} + \frac{T^{4} - (R - r_{c}')^{4}}{16 r'} \right]$$
(21)

where:

 $\alpha_{\rm g}$  = Total number of experimental alpha counts, dpm/particle.

 $\lambda$  = Specific alpha activity of uranium, dpm/µg.

 $N_{\parallel}$  = Concentration of uranium in migration zone,  $\mu g/\mu^3$ .

$$N_{U} = \frac{W_{U} P_{C} P_{U}}{W_{U} (P_{C} - P_{U}) + P_{U}} \times 10^{-6}$$

W = Weight fraction of uranium in migration zone.

 $p_{..}$  = Density of uranium, g/cm<sup>3</sup>.



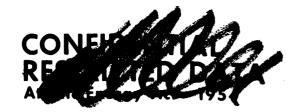


### **ACKNOWLEDGEMENT**

This report was prepared as a part of the work being performed under the Westinghouse Electric Corporation Subcontract NP-1 (NERVA Nuclear Subsystem) for the Aerojet-General Corporation Contract SNP-1 (NERVA Engine) under the auspices of the Space Nuclear Propulsion Office, a joint office of the Atomic Energy Commission and the National Aeronautics and Space Administration.

X66 50472







 $p_c$  = Density of pyrocarbon coat,  $g/cm^3$ .

Experimental counting efficiency factor.

 $r_c'$  = Range of an alpha particle in pyrocarbon coat,  $\mu$ .

- 23 -

 $R = Radius of coated fuel particle, \mu.$ 

 $T = Radial extent of migration, \mu.$